A hybrid particle-continuum method for hydrodynamics of complex fluids

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Outline

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Micro- and nano-hydrodynamics

- Flows of fluids (gases and liquids) through micro- (μm) and nano-scale (nm) structures has become technologically important, e.g., **micro-fluidics, microelectromechanical systems (MEMS).**
- **Biologically-relevant** flows also occur at micro- and nano- scales.
- The flows of interest often include **suspended particles**: colloids, polymers (e.g., DNA), blood cells, bacteria: **complex fluids**.
- Essential distinguishing feature from "ordinary" CFD: **thermal fluctuations**!

Example: DNA Filtering

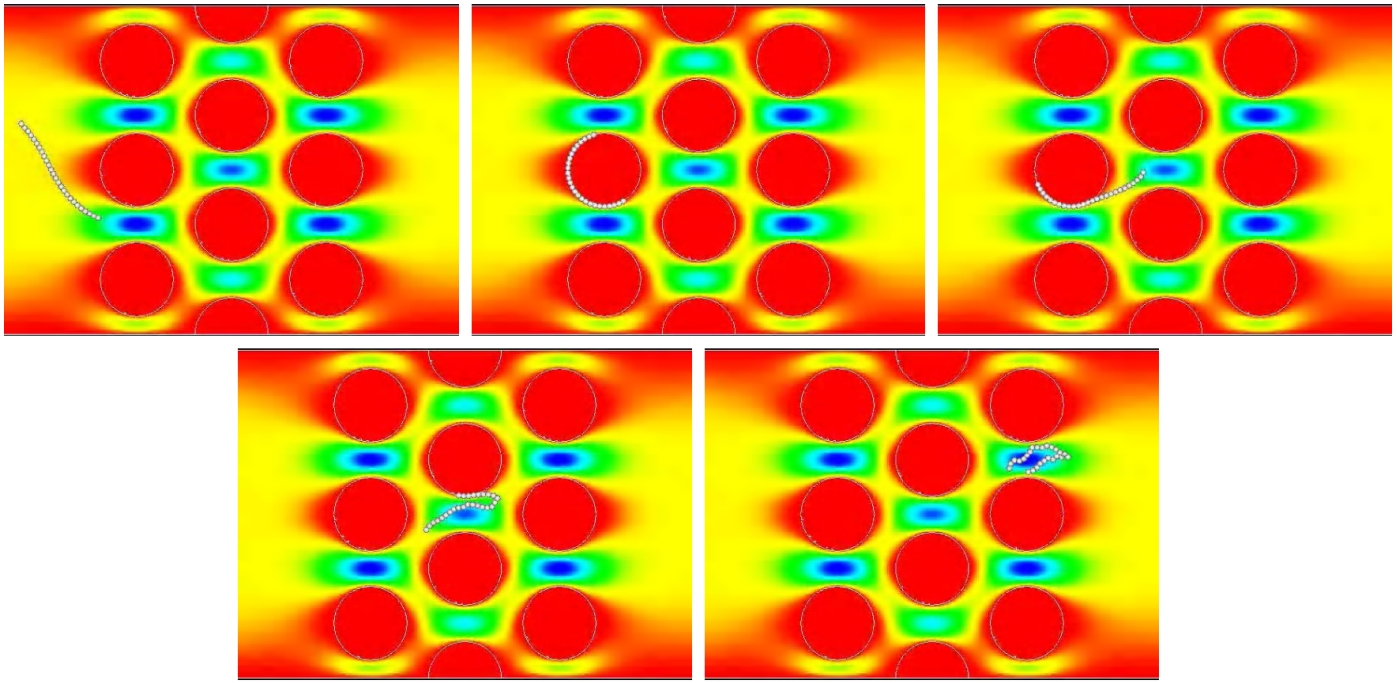


Figure: From the work of David Trebotich (LLNL)

Example: Droplet Formation

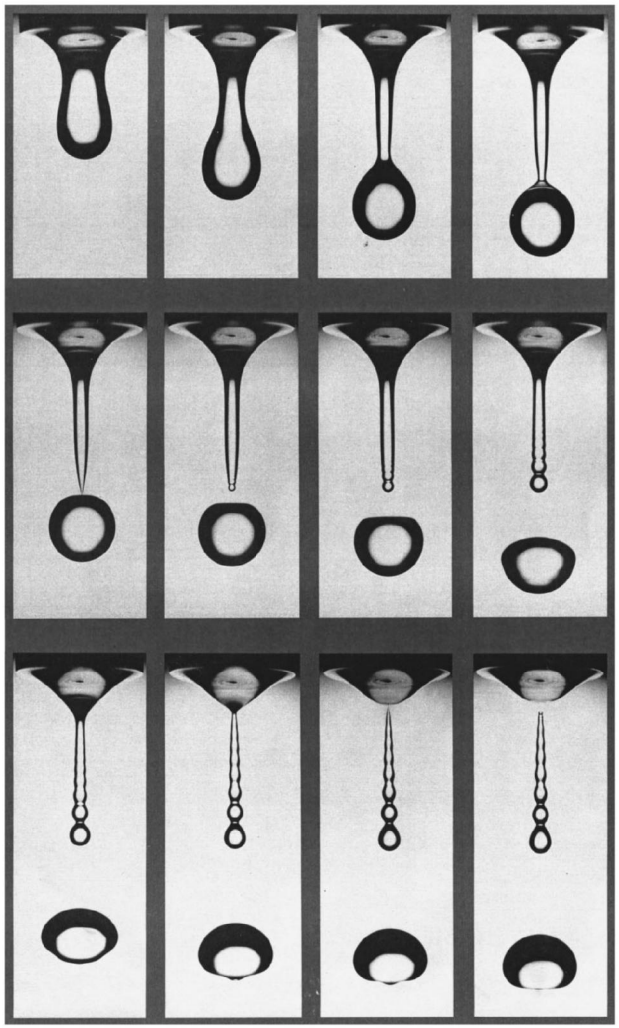
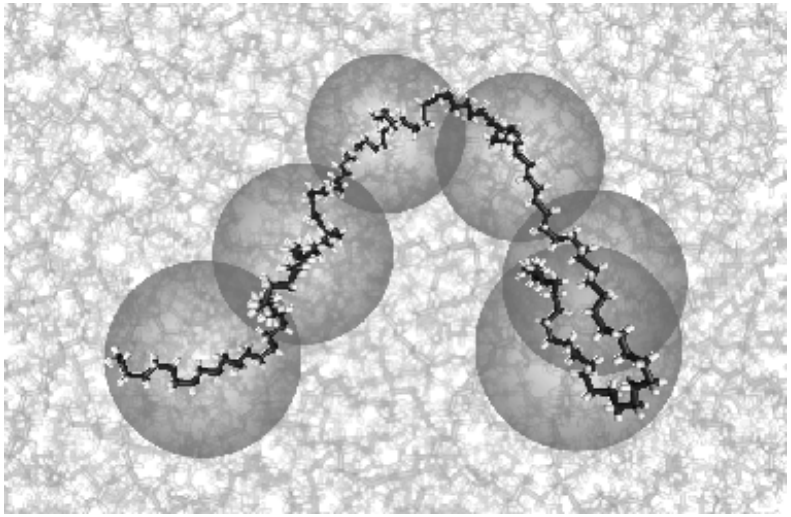


Figure: From Jens Eggers, Reviews of Modern Physics, 69, 1997

Polymer chains



Johan Padding, Cambridge

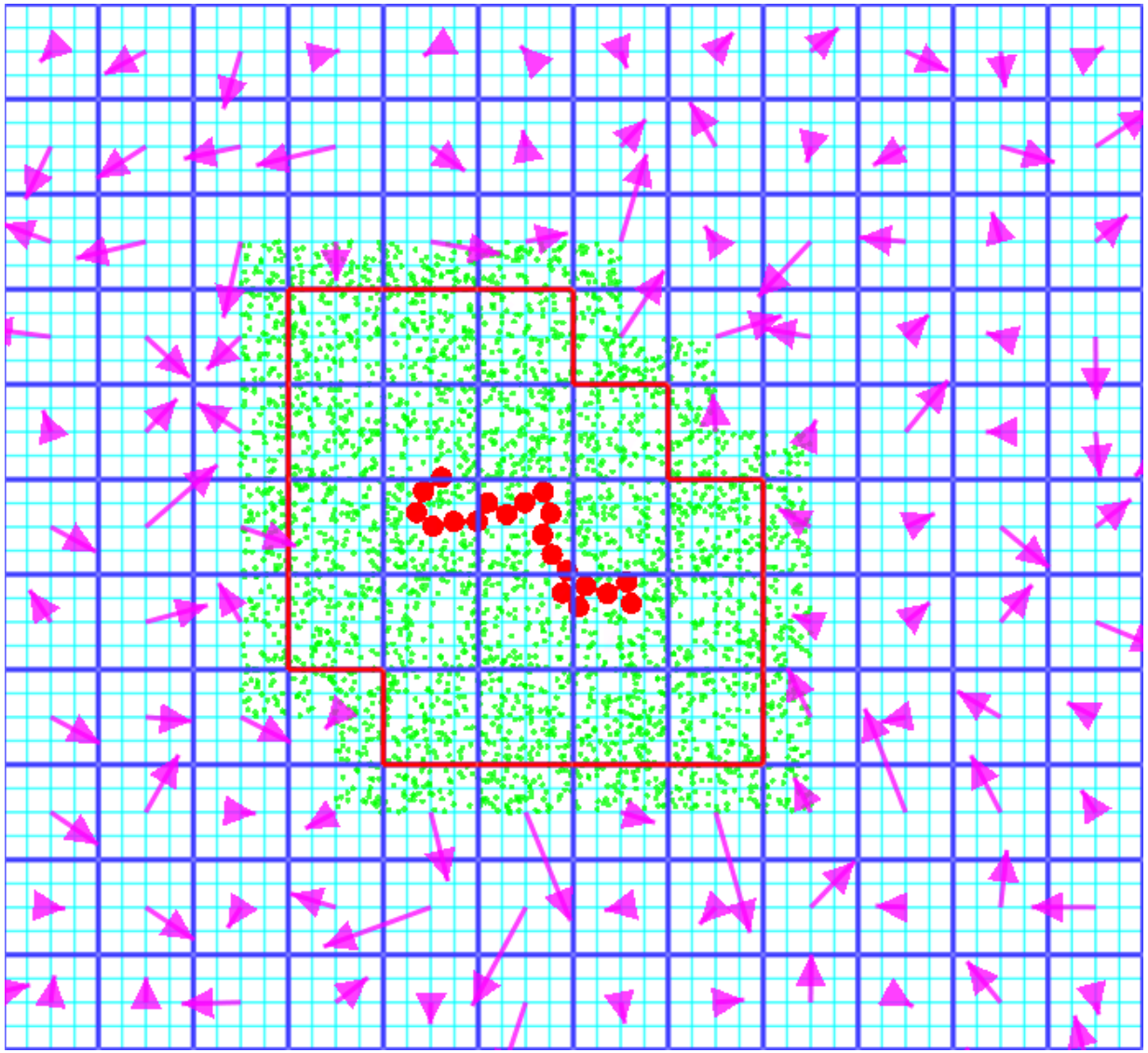
- I consider modeling of a polymer chain in a flowing solution, for example, DNA in a micro-array.
- The detailed structure of the polymer chain is usually **coarse-grained** to a model of spherical **beads**:

Bead-Link The beads are free joints between inextensible links

Bead-Spring Kuhn segments of the chain are point particles (beads) connected by non-linear elastic springs (FENE, worm-like, etc.)

The issue: **How to couple the polymer model with the surrounding fluid model?**

Particle/Continuum Hybrid Approach



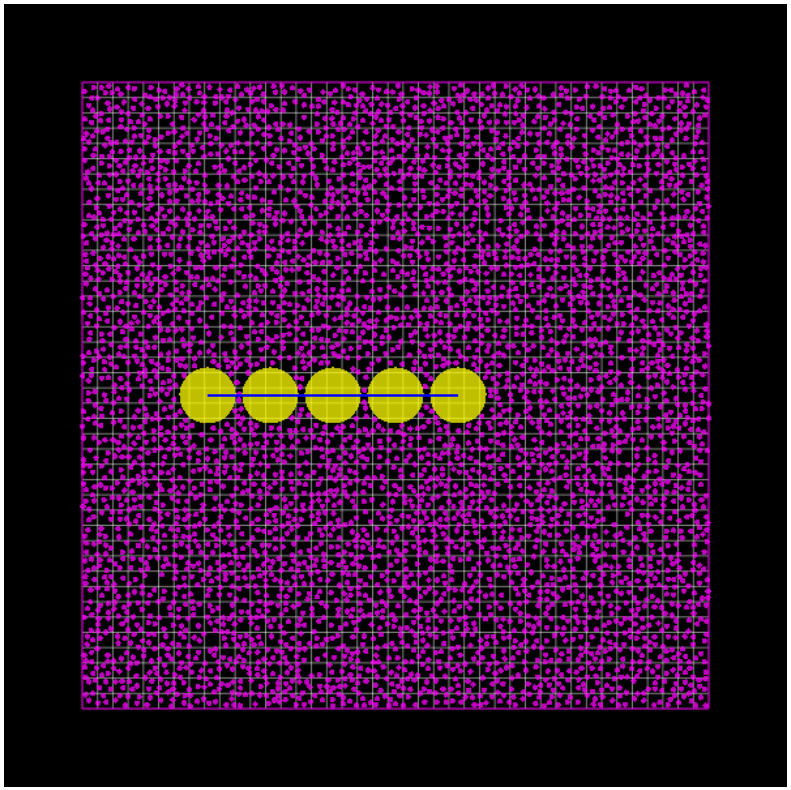
Particle Methods for Complex Fluids

- The most direct and accurate way to simulate the interaction between the **solvent** (fluid) and **solute** (beads, chain) is to use a particle scheme for both: **Molecular Dynamics (MD)**

$$m\ddot{\mathbf{r}}_i = \sum_j \mathbf{f}_{ij}(\mathbf{r}_{ij})$$

- Standard (time-driven) molecular dynamics:
  All of the particles are displaced *synchronously* in small *time steps* Δt, calculating positions and forces on each particle at every time step.
- The stiff repulsion among beads demands small time steps, and chain-chain crossings are a problem.
- For hard spheres, one can use **asynchronous event-driven MD**.
  "*Asynchronous Event-Driven Particle Algorithms*", by A. Donev, SIMULATION, 2009, **cs.OH/0703096**.

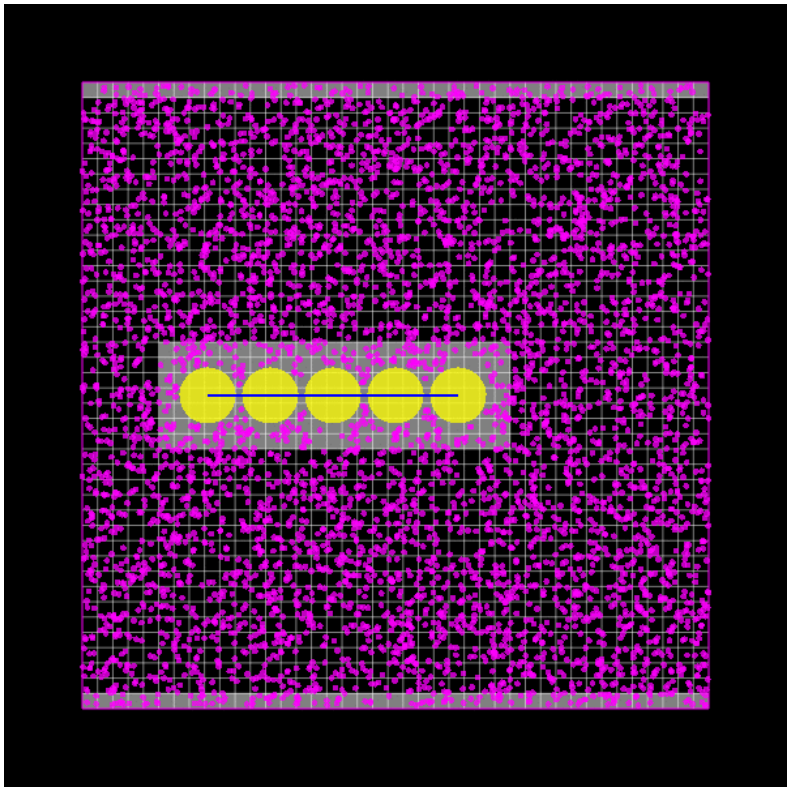
Event-Driven (Hard-Sphere) MD



- **Tethered** (square-well) hard-sphere chain polymers are the simplest but useful model.
- Most of the computation is "wasted" on the *unimportant solvent particles*!
- Over longer times it is **hydrodynamics** (*local momentum* and energy **conservation**) and **fluctuations** (Brownian motion) that matter.

(MNG)
"*Stochastic Event-Driven Molecular Dynamics*" [1],
A. Donev, A. L. Garcia and B. J. Alder,
J. Comp. Phys., 227(4):2644-2665, 2008

Direct Simulation Monte Carlo (DSMC)



- **Stochastic conservative collisions** of randomly chosen nearby solvent particles, as in Direct Simulation Monte Carlo (DSMC).
- Solute particles still interact with **both** solvent and other solute particles as hard spheres.
- Binary DSMC collisions can be replaced with **multiparticle collisions** (MPCD/SRD).

(MNG)

No fluid structure: Viscous ideal gas! [2]

"*Stochastic Hard-Sphere Dynamics for Hydrodynamics of Non-Ideal Fluids*", by A. Donev, A. L. Garcia and B. J. Alder, **Phys. Rev. Lett. 101:075902 (2008)** [arXiv:0803.0359, **arXiv:0908.0510**]

The Need for Coarse-Graining

- In order to examine the time-scales involved, we focus on a fundamental problem:
  A single bead of size a and density ρ' suspended in a stationary fluid with density ρ and viscosity η (**Brownian walker**).
- The isssue: Wide **separation of timescales occurs** *between the timescales of microscopic and macroscopic* processes as the bead becomes much bigger than the mean free path λ of the solvent particles.
- Typical bead sizes are *nm* (nano-colloids, short polymers) or μm (colloids, DNA), while typical atomistic sizes are $0.1nm$.

Estimates from Fluid Dynamics

- Classical picture for the following dissipation process: *Push a sphere suspended in a liquid with initial velocity* $V_{th} \approx \sqrt{kT/M}$, $M \approx \rho' a^3$, *and watch how the velocity decays:*
  - **Sound waves** are generated from the sudden compression of the fluid and they take away a fraction of the kinetic energy during a **sonic time** $t_{sonic} \approx a/c$, where c is the (adiabatic) sound speed.
  - **Viscous dissipation** then takes over and slows the particle *non-exponentially* over a **viscous time** $t_{visc} \approx \rho a^2/\eta$, where η is the shear viscosity.
  - **Thermal fluctuations** get similarly dissipated, but their constant presence pushes the particle diffusively over a **diffusion time** $t_{diff} \approx a^2/D$, where $D \sim kT/(a\eta)$.

Timescale Estimates

- The mean collision time is $t_{coll} \approx \lambda / v_{th} \sim \eta / (\rho c^2)$, where the thermal velocity is $v_{th} \approx \sqrt{\frac{kT}{m}}$, for water

$$t_{coll} \sim 10^{-15} s = 1 fs$$

- The **sound time**

$$t_{sonic} \sim \begin{cases} 1ns \text{ for } a \sim \mu m \\ 1ps \text{ for } a \sim nm \end{cases}, \text{ with gap } \frac{t_{sonic}}{t_{coll}} \sim \frac{a}{\lambda} \sim 10^2 - 10^5$$

Estimates contd...

- **Viscous time** estimates

$$t_{visc} \sim \begin{cases} 1\mu s \text{ for } a \sim \mu m \\ 1ps \text{ for } a \sim nm \end{cases}, \text{ with gap } \frac{t_{visc}}{t_{sonic}} \sim \sqrt{C}\frac{a}{\lambda} \sim 1 - 10^3$$

- Finally, the **diffusion time** can be estimated to be

$$t_{diff} \sim \begin{cases} 1s \text{ for } a \sim \mu m \\ 1ns \text{ for } a \sim nm \end{cases}, \text{ with gap } \frac{t_{diff}}{t_{visc}} \sim \frac{a}{\phi R} \sim 10^3 - 10^6$$

which can now reach **macroscopic timescales**!

Levels of Coarse-Graining

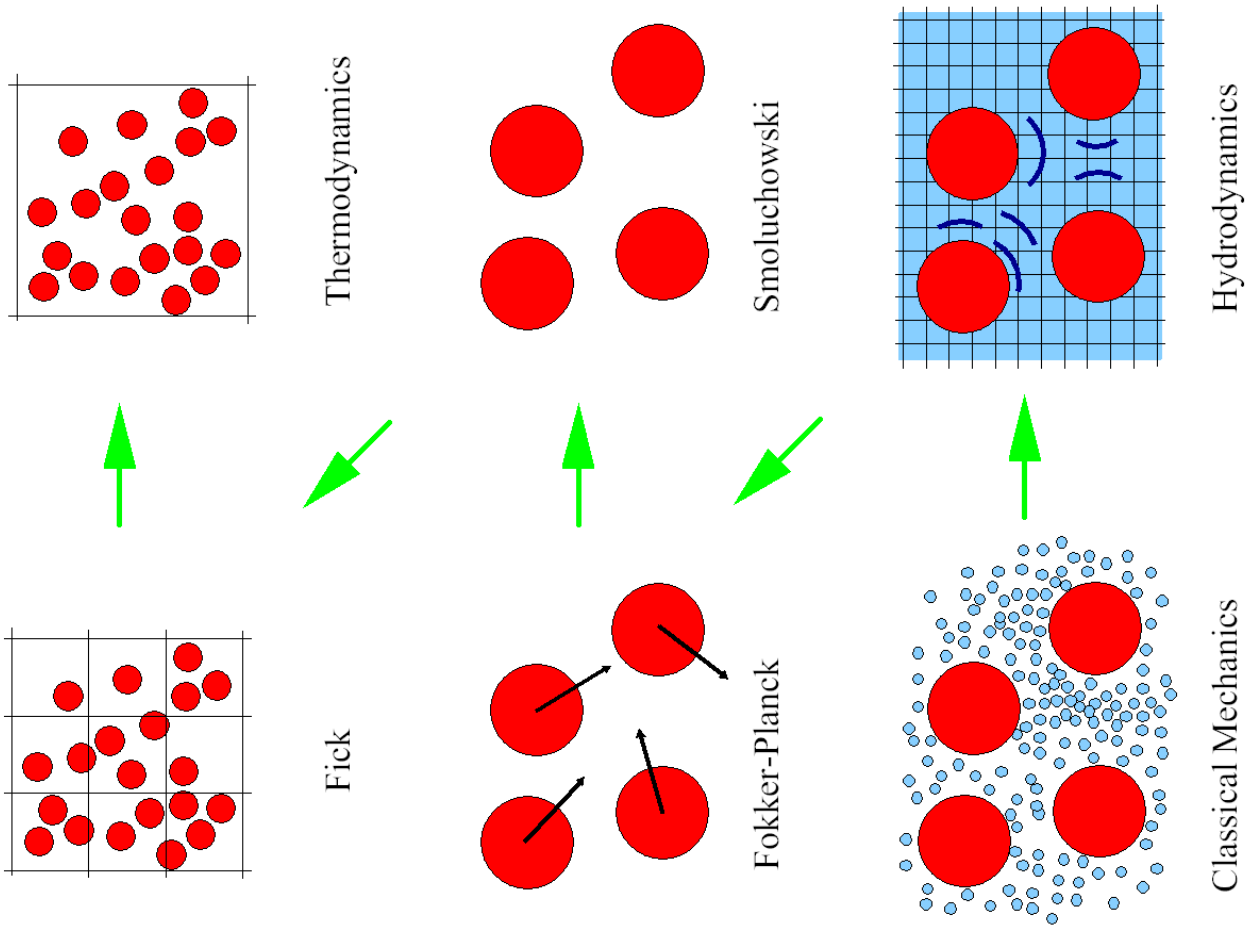


Figure: From Pep Español, "Statistical Mechanics of Coarse-Graining"

The equations of hydrodynamics

- Formally, we consider the continuum field of **conserved quantities**

$$\mathbf{U}(\mathbf{r},t) = \begin{bmatrix} \rho \\ \mathbf{j} \\ e \end{bmatrix} \cong \widetilde{\mathbf{U}}(\mathbf{r},t) = \sum_i \begin{bmatrix} m_i \\ m_i \mathbf{v}_i \\ m_i v_i^2/2 \end{bmatrix} \delta\left[\mathbf{r} - \mathbf{r}_i(t)\right],$$

  where the symbol $\cong$ means that $\mathbf{U}(\mathbf{r},t)$ approximates the true atomistic configuration $\widetilde{\mathbf{U}}(\mathbf{r},t)$ over long length and time scales.
- Due to the **microscopic conservation** of mass, momentum and energy,

$$\partial_t \mathbf{U} = -\boldsymbol{\nabla} \cdot \left[\mathbf{F}(\mathbf{U}) - \boldsymbol{\mathcal{Z}}\right] = -\boldsymbol{\nabla} \cdot \left[\mathbf{F}_H(\mathbf{U}) - \mathbf{F}_D(\boldsymbol{\nabla}\mathbf{U}) - \mathbf{B}\boldsymbol{\mathcal{W}}\right],$$

  where the flux is broken into a **hyperbolic, diffusive, and a stochastic flux**.
- Here $\boldsymbol{\mathcal{W}}$ is spatio-temporal white noise, i.e., a Gaussian random field with covariance

$$\left\langle \boldsymbol{\mathcal{W}}(\mathbf{r},t)\boldsymbol{\mathcal{W}}^{\star}(\mathbf{r}',t')\right\rangle = \delta(t-t')\delta(\mathbf{r}-\mathbf{r}').$$

Landau-Lifshitz Navier-Stokes (LLNS) Equations

Complete single-species fluctuating hydrodynamic equations:

$$\mathbf{U}(\mathbf{r},t) = \left[\ \rho,\quad \mathbf{j},\quad e\ \right]^T = \left[\ \rho,\quad \rho\mathbf{v},\quad c_v\rho T + \frac{\rho v^2}{2}\ \right]^T$$

$$\mathbf{F}_H = \begin{bmatrix} \rho\mathbf{v} \\ \rho\mathbf{v}\mathbf{v}^T + P(\rho,T)\,\mathbf{I} \\ (e+P)\mathbf{v} \end{bmatrix},\ \mathbf{F}_D = \begin{bmatrix} \mathbf{0} \\ \boldsymbol{\sigma} \\ \boldsymbol{\sigma}\cdot\mathbf{v} + \boldsymbol{\xi} \end{bmatrix},\ \mathcal{Z} = \begin{bmatrix} \mathbf{0} \\ \boldsymbol{\Sigma} \\ \boldsymbol{\Sigma}\cdot\mathbf{v} + \boldsymbol{\Xi} \end{bmatrix}$$

$$\boldsymbol{\sigma} = \left[\eta(\boldsymbol{\nabla}\mathbf{v} + \boldsymbol{\nabla}\mathbf{v}^T) - \frac{\eta}{3}(\boldsymbol{\nabla}\cdot\mathbf{v})\,\mathbf{I}\right] \text{ and } \boldsymbol{\xi} = \mu\boldsymbol{\nabla}T$$

$$\boldsymbol{\Sigma} = \sqrt{2k_B\bar{\eta}\overline{T}}\left[\mathcal{W}_T + \sqrt{\frac{1}{3}}\mathcal{W}_V\mathbf{I}\right] \text{ and } \boldsymbol{\Xi} = \sqrt{2\bar{\mu}k_B\overline{T}^2}\,\mathcal{W}_S$$

Problems with the LLNS equations

- Solving them numerically requires paying attention to **discrete fluctuation-dissipation balance**, in addition to the usual deterministic difficulties [3]!
- It is not clear whether the Navier-Stokes equations apply at **nano-scales**.
- Adding stochastic fluxes to the **non-linear** NS equations produces **ill-behaved stochastic PDEs**: At small scales one gets **negative densities** and **temperatures**.
- Mathematically-rigorous limit theorems only give the **linearized fluctuations** around the nonlinear mean, which lacks important physics.
- Fluctuations at scales smaller than the atomistic correlation length and time should be renormalized to account for discreteness of matter (recall *ultra-violet catastrophe*).

Spatio-Temporal Discretization

- Consider the general linear SPDE

$$\mathbf{U}_t = \mathbf{L}\mathbf{U} + \mathbf{K}\mathcal{W},$$

  where the **generator L** and the **filter K** are linear operators.
- Quite generally, numerical schemes use a **linear recursion** of the form

$$\mathbf{U}_j^{n+1} = (\mathbf{I} + \mathbf{L}_j \Delta t)\,\mathbf{U}^n + \Delta t \mathbf{K}_j \mathcal{W}^n = (\mathbf{I} + \mathbf{L}_j \Delta t)\,\mathbf{U}^n + \sqrt{\frac{\Delta t}{\Delta x}}\mathbf{K}_j \mathbf{W}^n$$

- For analysis, convert the iteration to Fourier space.
- When analyzing stochastic methods, it is natural to focus on the **second moments (covariance)**.

Stochastic Consistency and Accuracy

- Focus on the **discrete static spectrum**

$$\mathbf{S}_k = V \left\langle \widehat{\mathbf{U}}_k \left(\widehat{\mathbf{U}}_k \right)^\star \right\rangle = \mathbf{S}(k) + O\left(\Delta t^{p_1} k^{p_2}\right),$$

  for a **weakly consistent** scheme.
- Recall that $\mathbf{S}(\mathbf{k}) = \mathbf{I}$ for fluctuating conservation laws.
- The remainder term quantifies the *stochastic accuracy* for **large wavelengths ($\Delta k = k\Delta x \ll 1$) and small frequencies ($\Delta\omega = \omega\Delta t \ll 1$)**.
- A straightforward calculation **[3]** gives

$$\left(\mathbf{I} + \Delta t \widehat{\mathbf{L}}_k\right) \mathbf{S}_k \left(\mathbf{I} + \Delta t \widehat{\mathbf{L}}_k^\star\right) - \mathbf{S}_k = -\Delta t \widehat{\mathbf{K}}_k \widehat{\mathbf{K}}_k^\star.$$

Discrete Fluctuation-Dissipation

- For small Δt

$$\widehat{\mathbf{L}}_k \mathbf{S}_k^{(0)} + \mathbf{S}_k^{(0)} \widehat{\mathbf{L}}_k^\star = -\widehat{\mathbf{K}}_k \widehat{\mathbf{K}}_k^\star,$$

  and thus $\mathbf{S}_k^{(0)} = \lim_{\Delta t \to 0} \mathbf{S}_k = \mathbf{I}$ iff **discrete fluctuation-dissipation balance** [4] holds

$$\widehat{\mathbf{L}}_k + \widehat{\mathbf{L}}_k^\star = -\widehat{\mathbf{K}}_k \widehat{\mathbf{K}}_k^\star.$$

- Use the **method of lines**: first choose a spatial discretization consistent with the discrete fluctuation-dissipation balance condition, and then choose a temporal discretization.

"**On the Accuracy of Explicit Finite-Volume Schemes for Fluctuating Hydrodynamics**", by A. Donev, E. Vanden-Eijnden, A. L. Garcia, and J. B. Bell, 2009, submitted. [**arXiv:0906.2425**]

Three Dimensions

- In 3D, for **compressible flows**, the fluctuating velocities follow

$$\begin{aligned}\mathbf{v}_t &= \eta\left[\boldsymbol{\nabla}^2\mathbf{v}+\frac{1}{3}\boldsymbol{\nabla}\left(\boldsymbol{\nabla}\cdot\mathbf{v}\right)\right]+\sqrt{2\eta}\left[\left(\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{W}}_T\right)+\sqrt{\frac{1}{3}}\boldsymbol{\nabla}\mathcal{W}_V\right]\\ &= \eta\left(\mathbf{D}_T\mathbf{G}_T+\frac{1}{3}\mathbf{G}_V\mathbf{D}_V\right)\mathbf{v}+\sqrt{2\eta}\left(\mathbf{D}_T\boldsymbol{\mathcal{W}}_T+\sqrt{\frac{1}{3}}\mathbf{G}_V\mathcal{W}_V\right).\end{aligned}$$

- To obtain discrete fluctuation-dissipation balance, we require discrete **tensorial** divergence and gradient operators $\mathbf{G}_T=\mathbf{D}_T^\star$, and *vectorial* divergence and gradient $\mathbf{G}_V=\mathbf{D}_V^\star$.
- We use the **MAC** (marker-and-cell) discretizations for the tensorial operators and the **Fortin** (corner) discretization for vectorial operators, both previously used in incompressible projection schemes.
- For incompressible flows *only* the MAC discretization is required.

Fluid-Microstructure Coupling

- The **solvent** (fluid, liquid) can be modeled **implicitly** via analytical solutions (Brownian dynamics). But we want **reverse coupling** of the polymer motion on the flow (e.g., *drag reduction*)! We also need to resolve **shorter time scales** at nano systems.
- Macroscopically, the coupling between flow and moving bodies/structures/beads relies on:
  - **No-stick** boundary condition $\mathbf{v}_{rel} = 0$ at the surface of the bead.
  - Force on the bead is the integral of the stress tensor over the bead surface.
- The above two conditions are **questionable at nanoscales**, but even worse, they are very hard to implement numerically in an efficient and stable manner, even in the (phenomenological) Lattice-Boltzmann method.

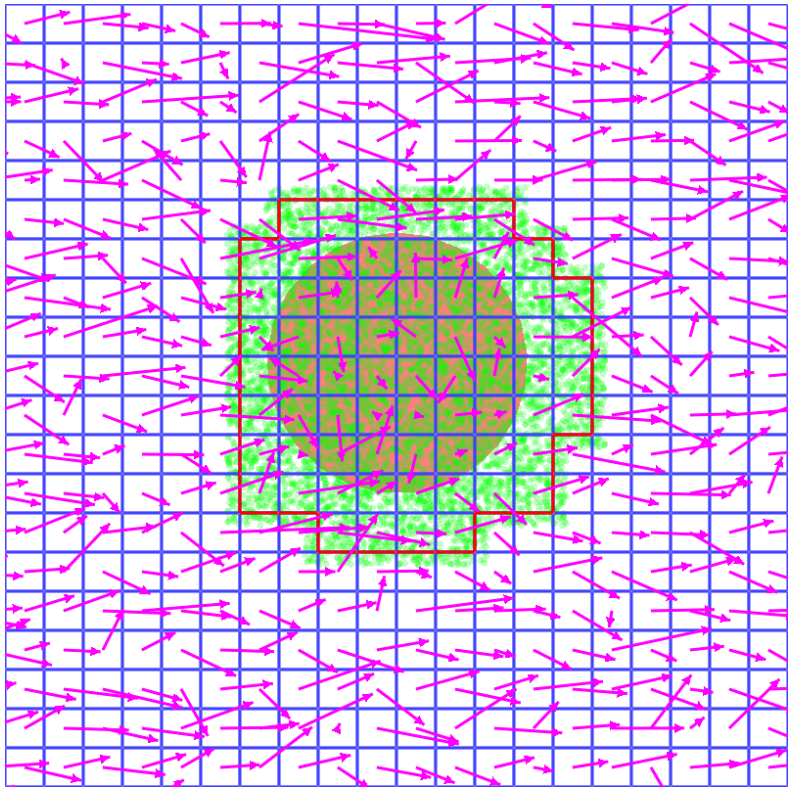
Point-Bead Approximations

- The **coupling** between the solute and solvent is **phenomenological and approximate** for most methods in use:

$$m d\dot{\mathbf{v}} = [\mathbf{F}(\mathbf{R}) - \gamma \mathbf{v}]\, dt + \sqrt{2\gamma k T}\, d\mathbf{W}$$

  - *Point beads* with artificial friction coefficients $\gamma \approx 6\pi a\eta$ based on asymptotic Stokes law
  - Point beads exerting (smeared) δ-function forces on the fluid
  - *Uncorrelated* fluctuating forces on the beads
- Such a **Langevin equation is physically inconsistent**, except at (unrealistic?) asymptotic time-scales!
- One can improve on this by **giving the beads a physical size** (not a point!) and consistently including **thermal fluctuations** in the fluid equations (see LB and SIB methods).

Solute-Solvent Coupling using Particles



Instantaneous (fluctuating) flow
Mean (plug) flow

- Split the domain into a **particle** and a **continuum (hydro) subdomains**, with timesteps $\Delta t_H = K \Delta t_P$.
- Hydro solver is a simple explicit **(fluctuating) compressible LLNS** code and is *not aware* of particle patch.
- The method is based on Adaptive Mesh and Algorithm Refinement (AMAR) methodology for conservation laws and ensures **strict conservation** of mass, momentum, *and* energy [5, 6].

Continuum-Particle Coupling

- Each macro (hydro) cell is either **particle or continuum**. There is also a **reservoir region** surrounding the particle subdomain.
- The coupling is roughly of the **state-flux** form:
  - The continuum solver provides *state boundary conditions* for the particle subdomain via reservoir particles.
  - The particle subdomain provides *flux boundary conditions* for the continuum subdomain.
- The fluctuating hydro solver is **oblivious** to the particle region: Any conservative explicit finite-volume scheme can trivially be substituted.
- The coupling is greatly simplified because the particle fluid is ideal (no internal structure): **No overlap region**.

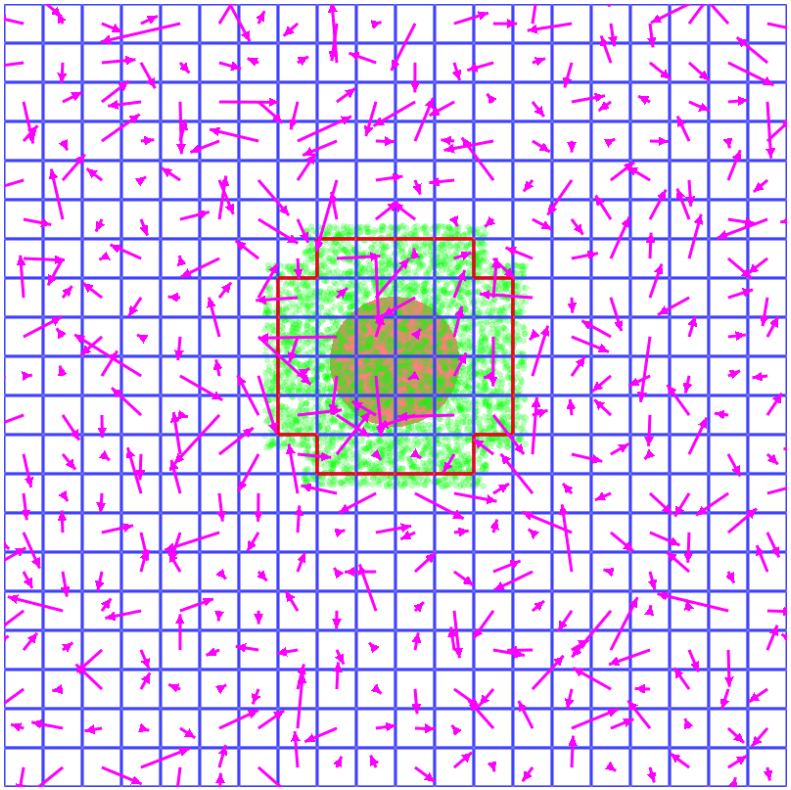
"**A hybrid particle-continuum method for hydrodynamics of complex fluids**" A. Donev and J. B. Bell and A. L. Garcia and B. J. Alder, to appear in SIAM Multiscale Modeling and Simulation, 2010.

Hybrid Algorithm

Steps of the coupling algorithm [7]:

1. The hydro solution is computed everywhere, including the **particle patch**, giving an estimated total flux $\mathbf{\Phi}_H$.
2. **Reservoir particles** are *inserted* at the boundary of the particle patch based on *Chapman-Enskog distribution* from kinetic theory, accounting for *both* collisional and kinetic viscosities.
3. Reservoir particles are *propagated* by Δt and *collisions* are processed (including virtual particles!), giving the total particle flux $\mathbf{\Phi}_p$.
4. The hydro solution is overwritten in the particle patch based on the particle state $\mathbf{u}_p$.
5. The hydro solution is corrected based on the more accurate flux, $\mathbf{u}_H \leftarrow \mathbf{u}_H - \mathbf{\Phi}_H + \mathbf{\Phi}_p$.

Back to the Brownian Bead



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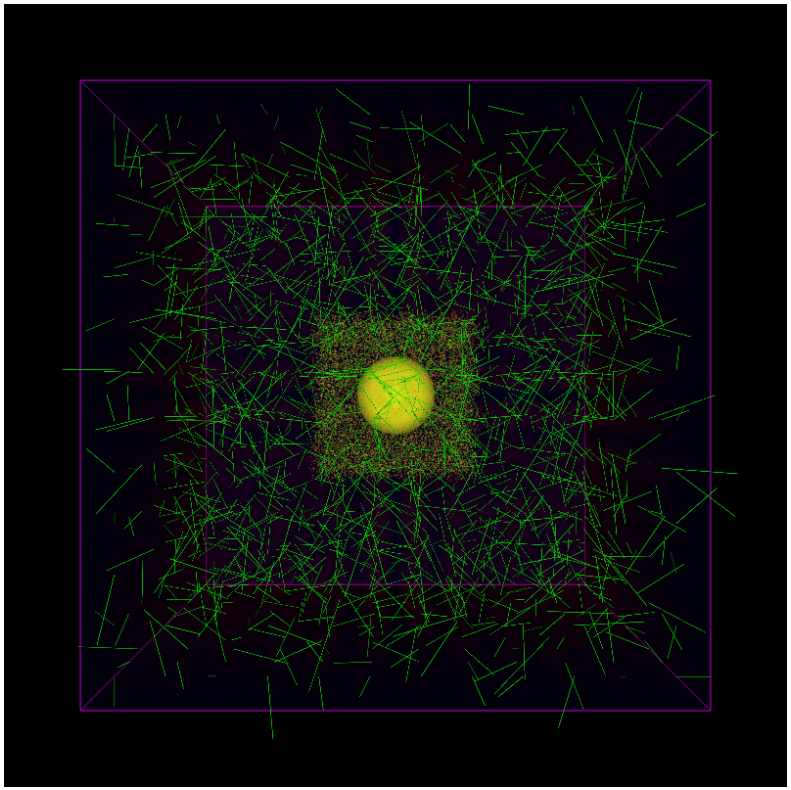
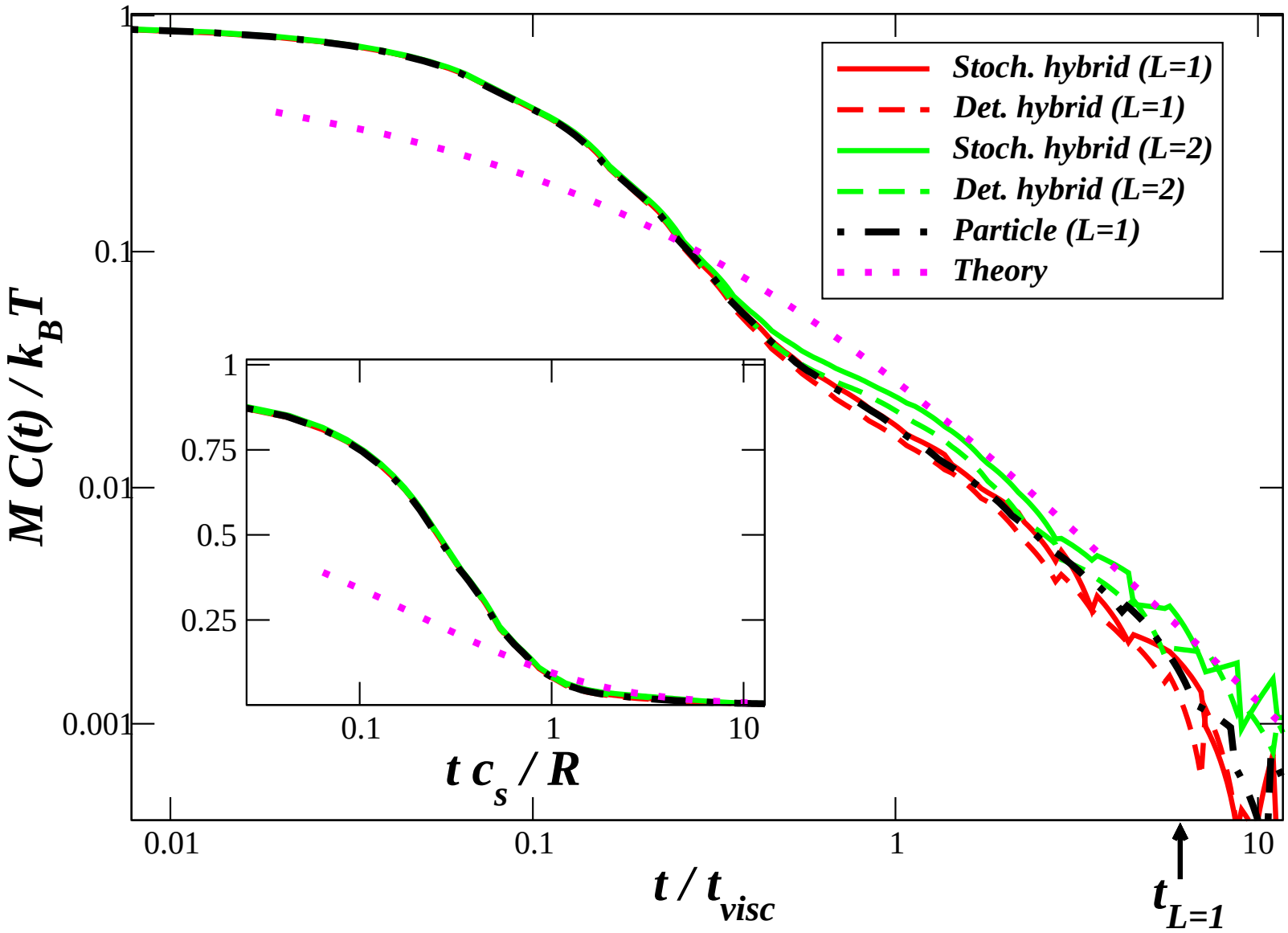


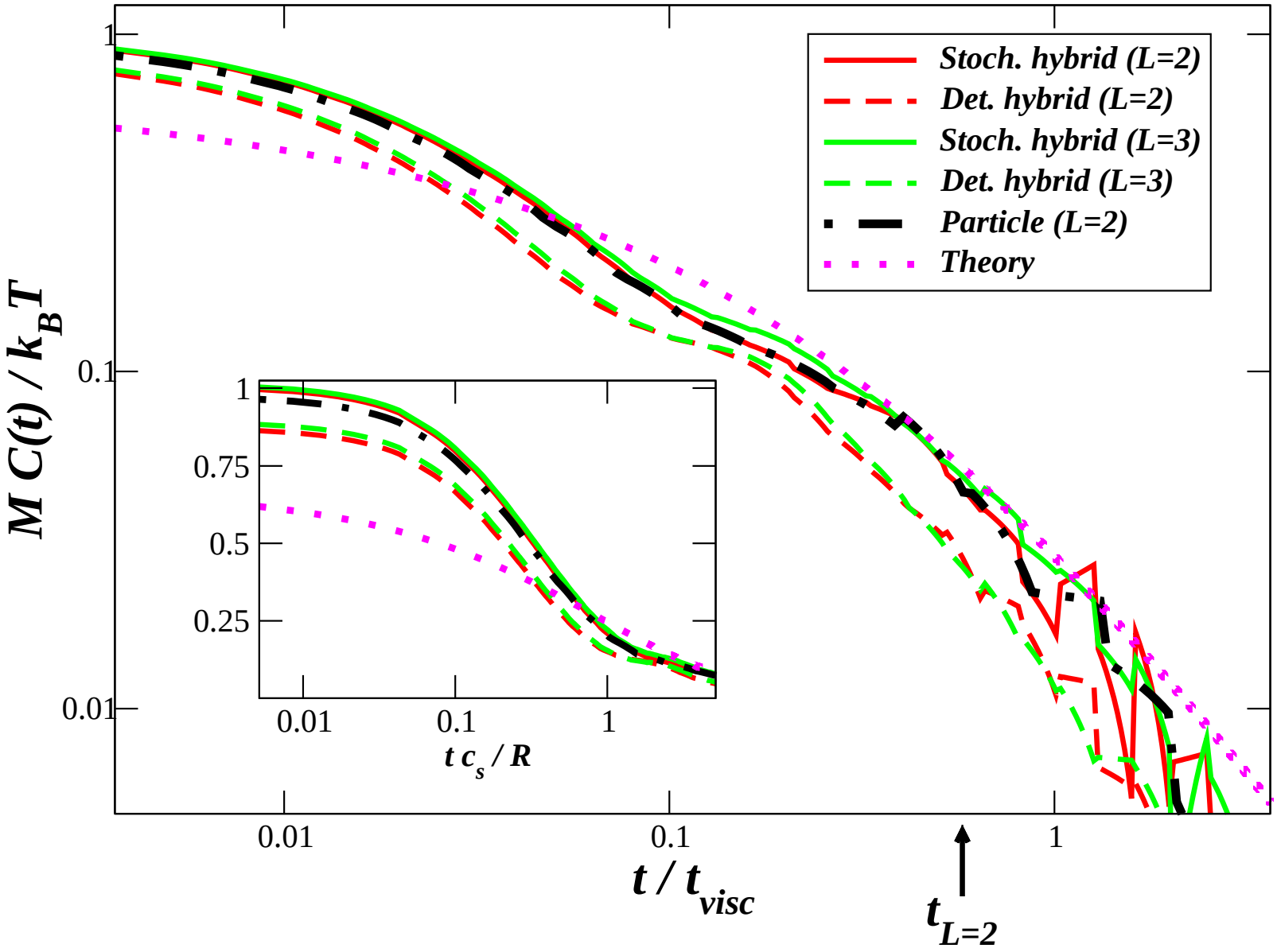
Velocity Autocorrelation Function

- We investigate the **velocity autocorrelation function** (VACF) for a Brownian bead
$$C(t) = \langle \mathbf{v}(t_0) \cdot \mathbf{v}(t_0 + t) \rangle$$
- From equipartition theorem $C(0) = kT/M$.
- For a **neutrally-boyant** particle, $\rho' = \rho$, incompressible hydrodynamic theory gives $C(0) = 2kT/3M$ because the momentum correlations decay instantly due to sound waves.
- Hydrodynamic persistence (conservation) gives a **long-time power-law tail** $C(t) \sim (kT/M)(t/t_{visc})^{-3/2}$ *not* reproduced in Brownian dynamics.

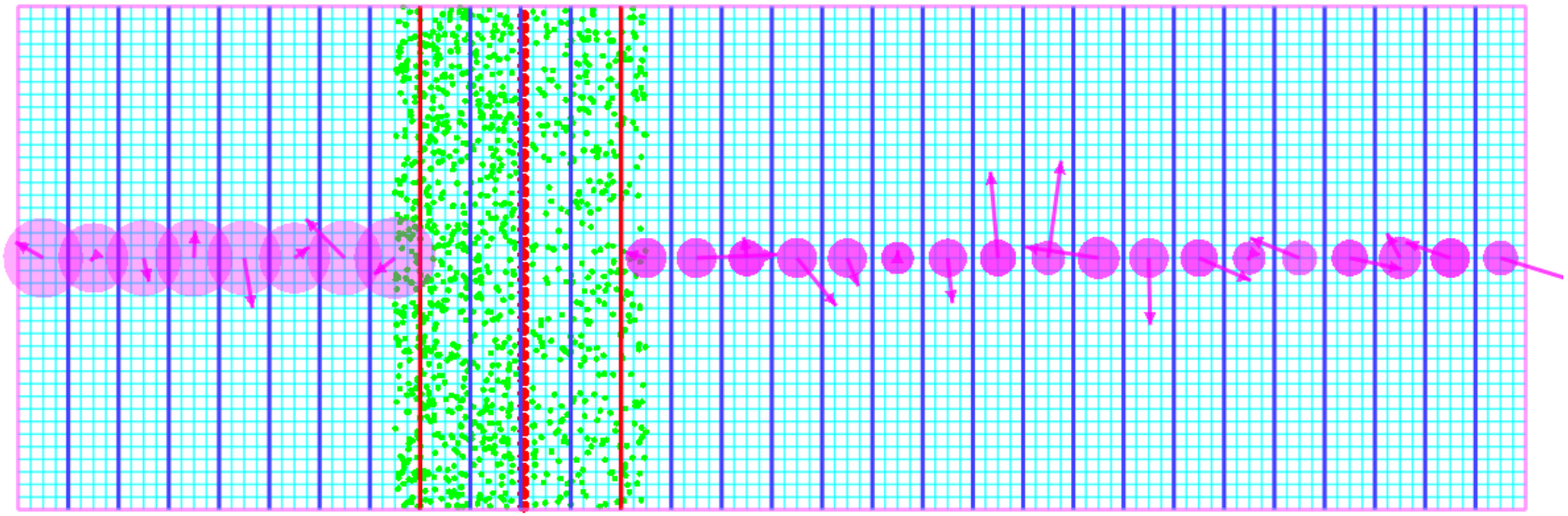
Small Bead (~10 particles)



Large Bead (~1000 particles)



The adiabatic piston problem



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Relaxation Toward Equilibrium

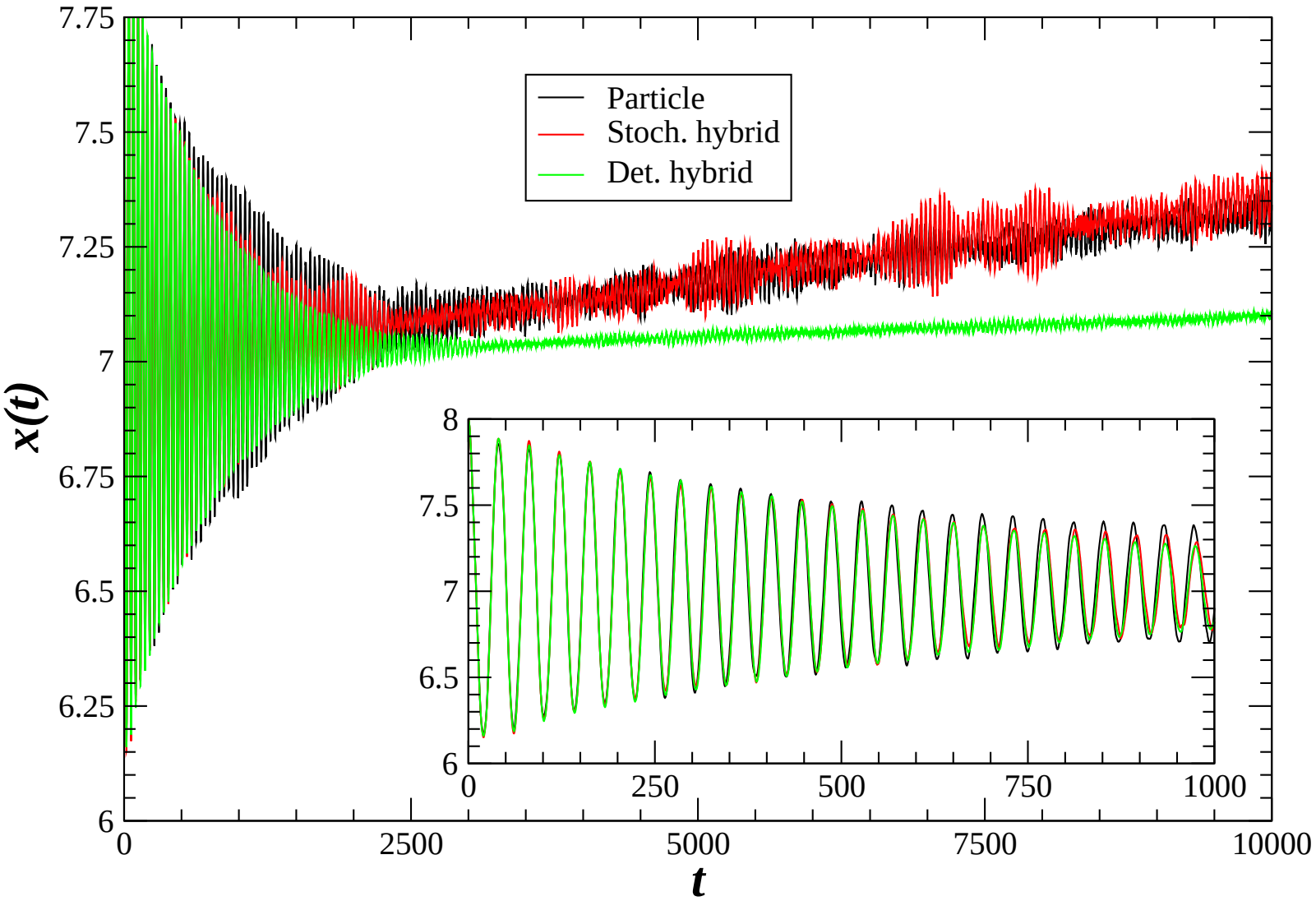

Figure: Massive rigid piston ($M/m = 4000$) not in mechanical equilibrium.

VACF for Piston

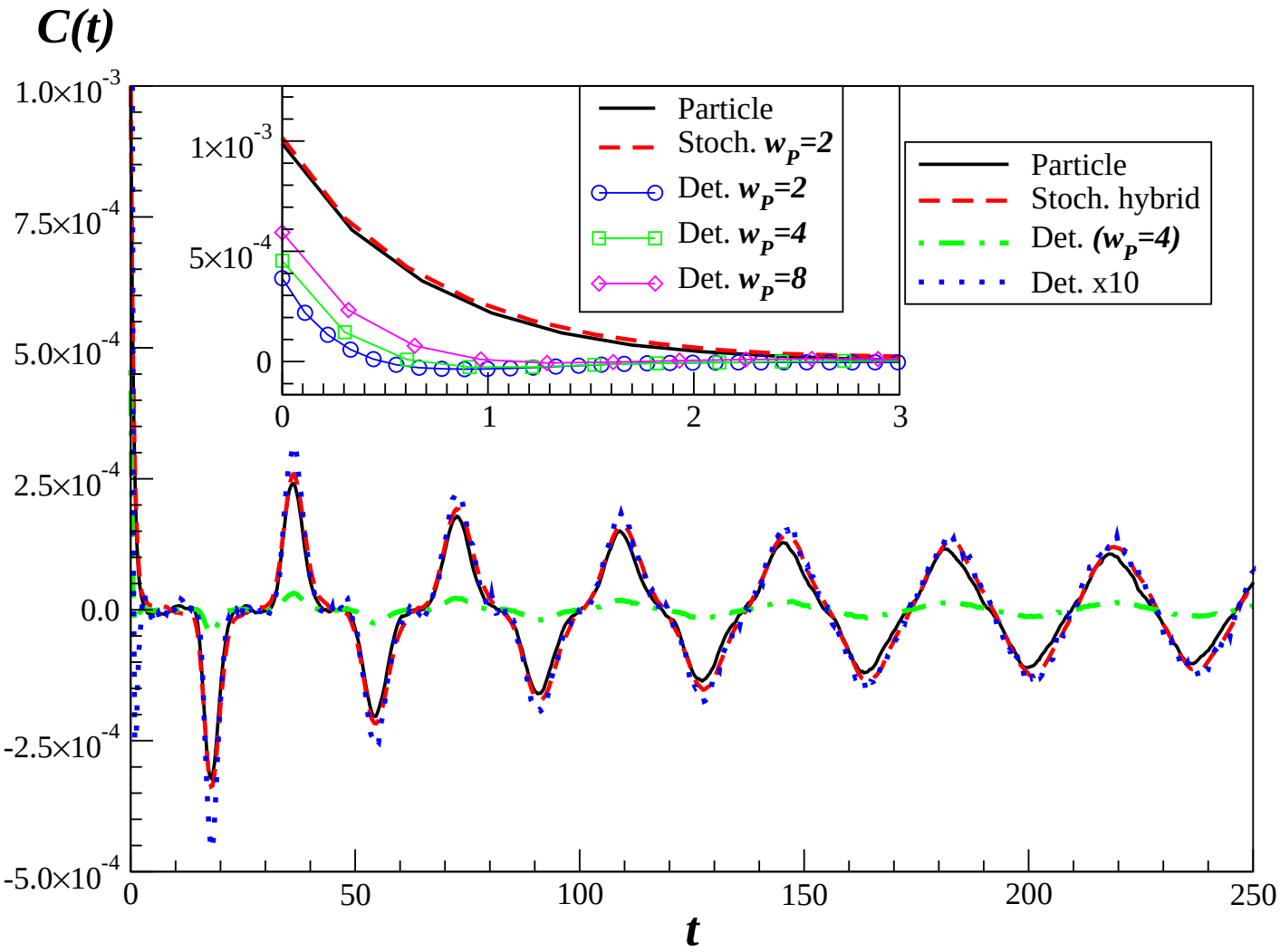

Figure: The VACF for a rigid piston of mas $M/m = 1000$ at thermal equilibrium.

Conclusions

- **Coarse-grained particle methods** can be used to accelerate hydrodynamic calculations at small scales.
- Designing numerical methods for fluctuating hydrodynamics requires attention to **fluctuation-dissipation balance**, in addition to the usual (deterministic) stability and accuracy considerations.
- Hybrid particle continuum methods closely reproduce purely particle simulations at a **fraction of the cost**.
- It is **necessary to include fluctuations** in the continuum subdomain in hybrid methods.

Future Directions

- Numerical schemes for **Low-Mach Number** fluctuating hydrodynamics.
- Theoretical work on the **equations of fluctuating hydrodynamics**: systematic coarse-graining and approximations.
- **Direct coupling** between fluctuating hydrodynamics and microstructure (solute beads).
- **Test, validate, and apply** the methodology for polymer problems.
- Couple our **non-ideal stochastic hard-sphere gas** to continuum hydrodynamics with *microscopic fidelity*.
- Ultimately we require an **Adaptive Mesh and Algorithm Refinement** (AMAR) framework that couples deterministic MD for the polymer chains (**micro**), a stochastic solvent (**micro-meso**), with compressible fluctuating Navier-Stokes (**meso**), and incompressible CFD (**macro**).

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